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AEROSOL NUMBER SIZE DISTRIBUTION IN THE BOREAL ENVIRONMENT: SPATIO-TEMPORAL VARIATION

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Academic dissertation

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Abstract

Atmospheric aerosols have an impact on the global radiation budget, and thus on climate, they reduce the air quality and visibility, and have multiple harmful health effects. The climatic significance of aerosols result from their ability to scatter and absorb solar radiation, and, if being large enough, mediate the cloud albedo and lifetime by acting as cloud condensation nuclei (CCN). The climatic effect, however, has a notable uncertainty.

Particles can be either directly emitted to atmosphere or they can form there from precursor vapors. The latter is called new particle formation (NPF). Globally, NPF has been estimated to be responsible for even half of CCN sized tropospheric particles. The understanding of the NPF mechanisms and the spatial and temporal variation of NPF in many scales is necessary to correctly represent aerosols in climate models.

In this work, we quantified the importance of biogenic organic vapours and anthropogenic sulfuric emissions in the NPF in northern boreal environment. Aerosol number size distribution data from three measurement sites were used to calculate the average continuous increase in aerosol particle diameter and number concentration when air masses travelled over land. A 14-year-long time series of aerosol and gas measurements were used to determine the effect of reduced Kola Peninsula SO_2 emissions on aerosol population at Eastern Finnish Lapland.

Secondly, this thesis describes in-situ aerosol measurements conducted with a light aircraft within the lowest 4 km of the troposphere. The data were used to determine the vertical and horizontal extent and variability of the NPF events in the surroundings of the Hyytiälä SMEAR II station. The airborne and ground level measurements were compared to find out the representativeness of the on ground measurements in the lowest parts of the atmosphere, in the planetary boundary layer.

The results showed that the Aitken mode particles grew, on avarage, at the apparent rate of around 1 nm h^{-1} when they travelled over the northern boreal environment during the growing season. The average calculated growth rates during the NPF events were 3–6 times higher than this apparent averege growth rate. The result implied that the condensation has a significant role in the particle growth even when NPF is not explicit. Also, the NPF events inside the planetary boundary layer were found to occur in area over a hundred kilometer. However, within this area, a notable variation in nucleation mode particles was observed.

Keywords: atmospheric aerosol, new particle formation, boreal environment, particle number size distribution, aircraft-borne measurements

Contents

List of publications

This thesis consists of an introductory review, followed by 5 research articles. In the introductory part, these papers are cited according to their roman numerals. **Papers I, II** and **V** are reprinted under the Creative Commons Licence. **Papers III** and **IV** are reproduced with the kind permission of the journals concerned.

I **Väänänen, R.**, Kyrö, E.-M., Nieminen, T., Kivekäs, N., Junninen, H., Virkkula, A., Dal Maso, M., Lihavainen, H., Viisanen, Y., Svenningsson, B., Holst, T., Arneth, A., Aalto, P. P., Kulmala, M. and Kerminen, V.-M.: *Analysis of particle size distribution changes between three measurement sites in northern Scandinavia*, Atmos. Chem. Phys., 13, 11887-11903, 10.5194/acp-13-11887-2013, 2013.

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1 Introduction

The atmosphere protecting us from the hostile space is an aerosol: it consists of solid and liquid particles suspended by gas. In case of the atmosphere, this gas consist of N_2, O_2, Argon and trace amounts of other gases. At the Earth's surface, aerosol particles cover only a mass fraction of up to 10^{-6} of carrier gas (Hinds, 1999), but they are tightly connected to two fundamental problems that humans have to solve. In addition to having climatic impacts as a cooling agent for the global climate (Stocker et al., 2013), particulate pollution also has negative health effects by increasing the risk for numerous diseases, from exposure of respiratory infections to cardiovascular symptoms, even increasing the mortality (WHO, 2016). Additionally, aerosols impair air quality by reducing visibility.

Aerosols influence the global radiation budget and thus affect the climate (Boucher et al., 2013). They can directly scatter or absorb solar radiation. This mechanism is called aerosol-radiation interaction. All aerosol particles scatter solar radiation and cool the atmosphere, but some aerosol types, such as black carbon, mineral dust and some organic components, can also absorb radiation causing a warming effect. Moreover, the radiation scattered or absorbed by aerosol particles cools or warms the surrounding air of the aerosol, affecting the nearby relative humidity and temperature, and further the cloud properties. Indirectly, the aerosols affect the radiation budget by changing the properties of clouds. This is called the aerosol-cloud interaction. Aerosol particles larger than about 50–100 nm can act as cloud condensation nuclei (CCN), which are the seeds for cloud droplets (Kerminen et al., 2012). Changes in the CCN concentrations lead to changes in cloud albedo and lifetime. Finally, if black carbon or other absorbing aerosol particles are deposited onto snow or ice, they reduce the surface albedo, resulting in a warming effect (Flanner et al., 2007). It is estimated that about half of the warming radiative forcing of CO_2 is counteracted by the current net radiative forcing of aerosol particles, but the uncertainties are large and the situation might change in future (Stocker et al., 2013).

The effects of the aerosols on climate or health depend on the chemical and physical properties aerosol particles, which vary abundantly. In the ambient atmosphere, the number concentration of aerosol particles ranges from few particles cm^{-3} in the very clean Antarctic environment (Järvinen et al., 2013) up to more than 10^5 cm^{-3}particles in in high polluted cities (Kumar et al., 2014). The mass concentration can be as high

as 1000 μg m^{-3}, and the size range of aerosol particles covers five orders of magnitude from few nanometers up to around a hundred micrometers (Hinds, 1999; Seinfeld and Pandis, 2006). The majority of the number concentration is covered by the smallest (<100 nm) particles, whereas larger (>100 nm) particles dominate the aerosol mass concentration. Unlike for many inactive gas-phase pollutants in the atmosphere, such as CO_2, the lifetime of aerosols is short, from hours to weeks (Williams et al., 2002; Croft et al., 2014), and during that time many atmospheric processes can modify the particle population. This causes the distribution of aerosol particles to have large variations in many scales, both temporary and spatially. To accurately estimate the non-linear impacts of aerosols on, for example, global climate, detailed information on aerosol populations, processes affecting them, and atmospheric conditions is needed. The understanding of aerosol processes (e.g. vapor condensation onto the existing particles, coagulation, aerosol removal processes and potential for CCN) requires knowledge not only on the number and mass concentrations, but also on the chemical composition and the size distribution of the aerosol population, as well as on their spatio-temporal variation.

The atmospheric aerosols are often classified by their origin into anthropogenic and natural. Another way to categorise them is to look at the birth mechanism and separate them into primary particles that are directly emitted to the atmosphere, and to secondary particles which are formed in the atmosphere. The sources of primary particles include emissions from combustion and traffic, erosion or resuspension induced mineral dust, sea salt aerosol production from sea water droplets, black carbon from biomass burning, and biogenic primary particles, such as fragments of plants or pollen. The other source of aerosol particles is that suitable vapors form particles in the atmosphere by gas-to-particle conversion, where the initial clusters are first formed by nucleation and then grown in size by vapors condensing onto them. This process is called new particle formation (NPF, Kulmala et al., 2013; Zhang et al., 2012), and it has been observed in diverse environments around the world (Kulmala et al., 2004). Sulfuric acid is found to be the most important precursor vapor for NPF (Weber et al., 1995; Kulmala and Kerminen, 2008). The separation into natural and anthropogenic particles often fails for the secondary particles since the vapors can have multiple origins. The mechanisms related to nucleation and consequent growth depend on the available vapors, as well as on meteorological conditions and the sink for these vapors due to the pre-existing aerosol particles. These can vary spatially and temporally, by natural reasons or by anthropogenic causes.

In this thesis, aerosol number size distribution measurements both at ground stations and in-situ airborne were used to quantify the natural and anthropogenic factors affecting the new particle formation in the boreal environment. The main objectives of this thesis were

i) to quantify the particle growth due to condensation of vapors originating from emissions from the boreal environment in Northern Scandinavia, and its spatial variability between three measurement stations (**Paper I**);
ii) to resolve the effect of Kola Peninsula sulfur emissions on the particle population in Eastern Finnish Lapland and its evolution during a 14-year-long measurement period (**Paper II**).
iii) to implement airborne aerosol measurements as a part of an extensive field campaign (**Paper III**).
iv) to quantify the horizontal and vertical variation of the particle number size distribution in the lower atmosphere over boreal environment in order to understand the relevant spatial scales of the new particle formation (**Papers IV** and **V**).
v) to estimate the representativeness of particle number size distributions measured at the Hyytiälä SMEAR II station by comparing them to airborne measurements (**Paper V**)

2 Atmospheric processes

2.1 Atmospheric new particle formation

The atmospheric aerosol population is constantly evolving. Particles are emitted to the atmosphere as primary particles or they can nucleate in the atmosphere from suitable vapors as secondary particles. They can grow in size by condensation or coagulation, while wet or dry deposition can remove them from the atmosphere.

Atmospheric *new particle formation* (NPF) is a phase transition, where the precursor vapors collide to form small clusters of molecules, and these clusters subsequently grow to larger sizes by suitable vapors condensing onto them (Vehkamäki and Riipinen, 2012; Kulmala et al., 2014). NPF events are frequent almost everywhere on the planet (Kulmala et al., 2004; Kulmala and Kerminen, 2008; Manninen et al., 2010), from continental Antarctica (Järvinen et al., 2013) to high altitude mountains (Rose et al., 2015), and from urban cities (Yue et al., 2011) to rural background stations (Asmi et al., 2011, **Paper I, Paper II)**. It is estimated that even half of cloud condensation nuclei (CCN) concentration in the troposphere originates from NPF (Spracklen et al., 2008; Merikanto et al., 2009; Reddington et al., 2011).

Sulfuric acid is found to be the key component to drive the first steps of the NPF process at the lowermost part of the atmosphere in the planetary boundary layer (Weber et al., 1995, 1997; Kulmala et al., 2007, 2013; Petäjä et al., 2009; Sipilä et al., 2010). However, other vapors are needed to stabilize the sulfuric acid clusters. Recent studies have revealed in a molecular level that several compounds, or their mixtures, including ammonia (Kirkby et al., 2011; Lehtipalo et al., 2016), amines (Kúrten et al., 2008; Petäjä et al., 2011; Almeida et al., 2013; Lehtipalo et al., 2016), highly oxidized organic compounds (Kulmala et al., 1998; Schobesberger et al., 2013; Ehn et al., 2014; Riccobono et al., 2014) and ions (Kirkby et al., 2011), can have a role in the NPF process, but the understanding of the mechanisms is still limited. Recent studies have also reported NPF under conditions in the absence of sulfuric acid, such as in a laboratory chamber measurements (Tröstl et al., 2016; Kirkby et al., 2016) or in the pristine free troposphere (Bianchi et al., 2016). Then NPF was initiated by highly oxygenated molecules (HOMs), which are oxidation products of, for example, biogenic volatile organic compounds (Ehn et al., 2012, 2014). When the nucleated particles grow further in size, sulfuric acid concentrations are not sufficient alone to explain the observed

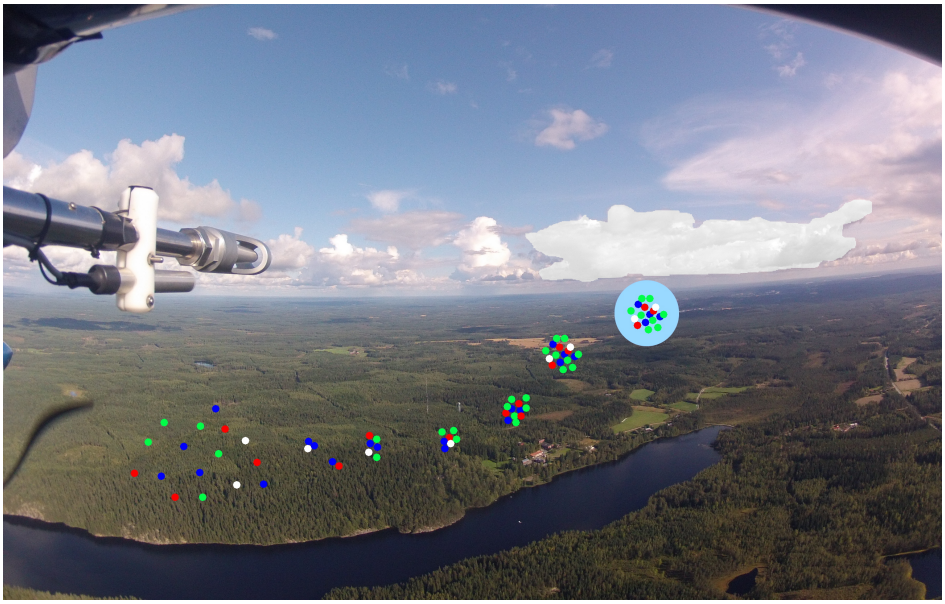

Figure 2.1: Vapors in trace concentrations can nucleate into particles which further can act as cloud condensation nuclei.

growth rates (Weber et al., 1995; Kulmala et al., 1998; Kirkby et al., 2011; Riipinen et al., 2012), and organics usually start to dominate the growth (Kulmala et al., 2013). Within favorable conditions, the vapors can continue to condense onto the clusters, and this subsequent growth of the particles can bring them up to sizes where they can act as CCNs and thus have a climatic significance (Fig. 2.1) (e.g. Kerminen et al., 2012).

When interpreting atmospheric measurements, one needs to keep in the mind that air masses are practically always advecting, and the observations at a fixed measurement station do not follow the growth of certain particles, but the Eulerian dynamics of the particle number size distribution is measured. However, when the properties of the air masses are regionally homogeneous, a continuous growth of the particles can be observed. When measuring the size distribution of aerosols as a function of time, regional-scale nucleation events are observed as a new sub-25 nm particle mode that grows for several hours. Due to the shape of these curves, they are called 'banana plots'. The NPF events can cover areas of tens or hundreds of kilometers (Vana et al., 2004, 2016; Dal Maso et al., 2005; Wehner et al., 2007; Hussein et al., 2009; Crippa and Pryor, 2013; Kristensson et al., 2014). When the air mass where NPF occurs is less homogeneous, i.e. there is more variation over the area in the background aerosol distribution, in the precursor gas species and emission concentrations, or in meteorological conditions, a less regular growth is observed at a fixed station. As an example,

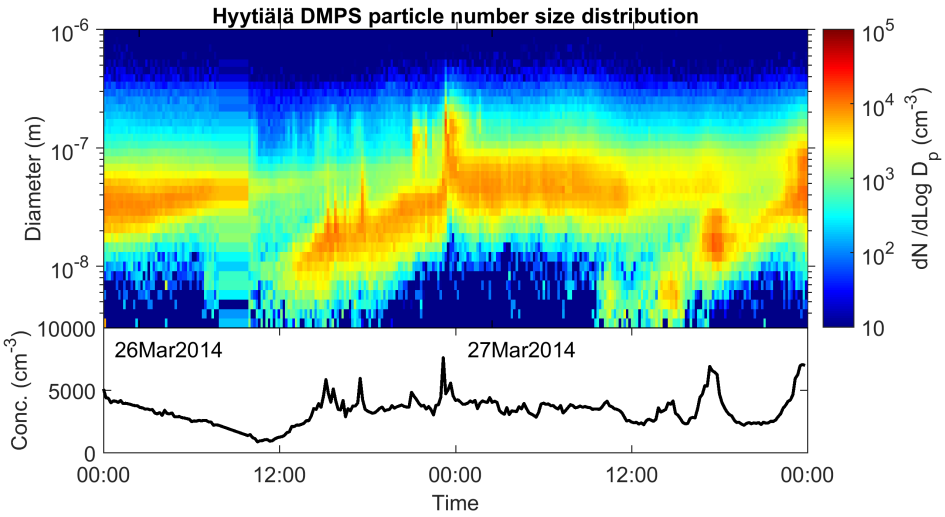

Figure 2.2: Two NPF events observed on concurrent days 26 and 27 March 2014 at SMEAR II station. The particle growth on the first day is smoother than during the later one.

Fig. 2.2 shows particle number size distribution and number concentration measured at the SMEAR II station on two consecutive days during an airborne measurement campaign in March 2014. The event on the first day had a smoother growth than observed on the second day.

The chemical compounds participating in the NPF and growth have different origins to the atmosphere. The main source of sulfuric acid is the oxidation process of sulfur dioxide (SO_2). SO_2 originates mainly from anthropogenic emissions but also for example from volcanic activity or dimethylsulfide emissions from oceans (Seinfeld and Pandis, 2006). The reduction of sulfur pollution during the last decades has had an effect on the aerosol concentrations (Chin et al., 2014, **Paper II)**. The sources of amines in the atmosphere include agriculture, biomass burning, oceans and vegetation (Ge et al., 2011). Organic molecules, which exist in numerous individual compounds, can participate in NPF after oxidation processes producing compounds with lower saturation pressure. They originate not only from different biological processes from terrestrial and ocean ecosystems, but also from many anthropogenic sources (Goldstein and Galbally, 2007).

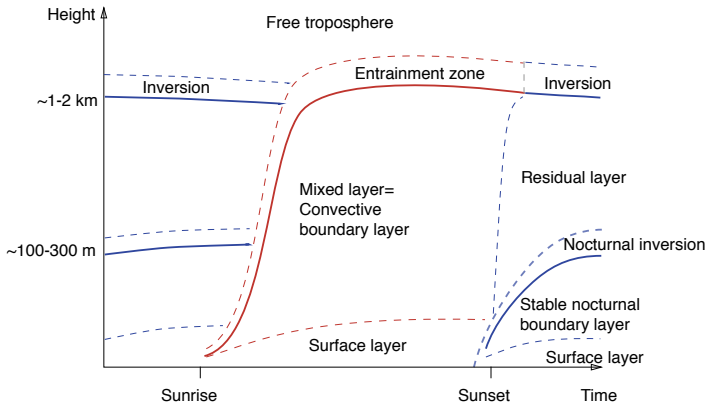

Figure 2.3: Schematic figure of diurnal evolution of the planetary boundary layer (adapted from Stull 2012 and Garratt 1992)

2.2 Planetary boundary layer evolution

The planetary boundary layer (PBL, also called atmospheric boundary layer, ABL, or just boundary layer, BL) is the lowermost part of the atmosphere. It is defined as the layer that is directly connected to the Earth's surface through the exchange of momentum, heat and mass, and responds to surface forcing within a timescale of an hour or less (Stull, 2012). The atmosphere above the PBL is called free troposphere (FT) or free atmosphere. Stull (2012) defines the bottom 10 % of the boundary layer as a surface layer. There the heat conduction, evaporation and frictional drag generate large vertical gradients in the wind speed, temperature, and humidity (Stull, 2000).

The continental PBL driven by thermal turbulence has a characteristic diurnal cycle (Fig. 2.3). Before the sunrise, the thermal turbulence has its minimum. The PBL consists of a stable nocturnal boundary layer (NBL) and residual layer (RL) above it. After the sunrise, the solar radiation starts to warm the Earth's surface, which results in heat flux up to atmosphere. This induces turbulent eddies which break the night time stable atmospheric stratification and lead to the mixing of the lowermost atmosphere. Also kinetic turbulence, i.e. wind shear, can affect the mixing of the lowermost atmosphere. The resulting rising layer is called the mixed layer (ML) or the convective boundary layer (CBL). At the top of the mixed layer, there is a thin entrainment zone, where first in the morning the residual layer, and later in the day the free troposphere, is entrained into the mixed layer and vice versa by the thermal turbulence. Typically there is a thermal inversion between the mixed layer and the free troposphere.

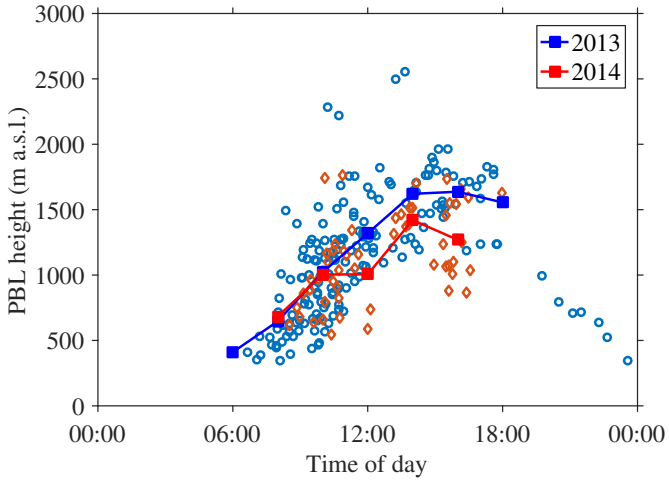

Figure 2.4: A composite picture of the PBL height develop near Hyytiälä as a function of time of day. Datapoints observed during aircraft-borne measurement campaigns on springs 2013 and 2014. Bold dots and curve show 2h average heights.

The convective turbulence dies out after sunset. The stable nocturnal boundary layer (NBL) starts to form and it grows during the night. Due to the radiation cooling of the ground, a nocturnal inversion layer is often formed above the surface. The residual layer above the NBL is a near adiabatic layer, which is no longer connected to the surface, and it often preserves the moisture from the day's mixed layer. Due to the lack of energy input from solar radiation, photo-chemical processes in the residual layer are damped.

The height of the fully developed PBL varies from day-to-day together with the season and meteorological conditions, and, in addition, has spatial variance caused by the surface topology and local meteorology. Fig. 2.4 shows how the ML height varied as a function of time of day during May–June 2013 and March–April 2014 aircraft-borne measurement campaigns. The temperature during the 2013 campaign was higher which induced a higher PBL height. Typically in Finland the PBL height rises up to 1–2 km, but it can rise above 2.5 km, as also our measurements during the spring campaign in 2013 showed (Fig. 2.4), or in the winter it can be as low as 300 m. The potential temperature and the water vapor mixing ratio have constant values inside the mixed PBL, but can have sharp gradients at the top of it. Their vertical profiles thus provide a method to detect the height of the PBL.

2.3 New particle formation inside the planetary boundary layer

The precursor gases for NPF (e.g. anthropogenic SO_2 which is oxidized to H_2SO_4, terpenoids from vegetation that also are further oxidized to HOMs) are emitted almost entirely from the surface level. In calm air, vapor concentrations tend to be highest at lowest altitudes and then gradually decrease vertically due to diffusion. In a study by O'Dowd et al. (2009), conducted by a small and a microlight aircraft over the background station Hyytiälä, nucleation mode particles were found first to peak at the lowest altitudes, just above the canopy top.

The mixing of air by turbulence and convection, and on the other hand limited mixing between the different atmospheric layers, makes the situation more complicated.

The onset of NPF at the ground level is observed to take place usually in the morning, concurrent with the growth of the mixed layer (Väkevä et al., 2000; Nilsson et al., 2001; Wehner et al., 2010; Altstädter et al., 2015; Platis et al., 2016). Solar radiation, which causes the morning development of the PBL, also promotes the photo-oxidation processes of the precursor vapors, and leads to an increased HOM production (Jimenez et al., 2009). They, in turn, participate in the nucleation and growth of particles. In addition to this, several processes related to the PBL development itself can affect nucleation. The observations have confirmed that a NPF inside the PBL is limited to the mixed, residual and entrainment layers (Stratmann et al., 2003; Wehner et al., 2007, 2010; Crumeyrolle et al., 2010; Platis et al., 2016, **Papers IV, V)**, but where exactly the nucleation takes place, is still under investigation.

After sunrise, solar radiation provides energy for the thermal turbulence, and the ML starts to rise. As a result, possibly cleaner air first from the RL and later from the free troposphere mixes with the air in the ML, and the existing aerosol concentration in the ML can thus dilute. This typically leads to a reduced condensation sink which favors NPF (Kulmala et al., 1998; Väkevä et al., 2000). Nilsson et al. (2001) suggested that there can be different precursor vapor concentrations in the ML and the RL. If there are enough vapors to form initial clusters but not sufficiently suitable (organic) vapors for the growth of the particles in the RL, the mixing of the RL air downwards to the ML could create more favorable conditions for NPF. Stratmann et al. (2003) and Platis et al. (2016) observed increasing concentrations of 5–10 nm particles in the RL before the break-up of the nocturnal inversion while nucleation mode particles on-ground were

observed shortly after it. Wehner et al. (2007) found that the NPF started separately at different altitudes both in the ML and the RL.

The atmospheric turbulence by convection is suggested to enhance the NPF rate by two mechanisms. First, updrafts can lift up precursors to higher altitudes, where colder environment is shown to promote nucleation (Easter and Peters, 1994). These updrafts can be connected to the mesoscale PBL circulation, such as roll vortices (Buzorius et al., 2001; Lampilahti, 2016). Second, in cases where the mean concentrations of the precursor gases are too low for the NPF to occur, the local fluctuations in the temperature and humidity can create 'pockets' with supersaturation of precursor gases (Nilsson and Kulmala, 1998). Observations have found small particles within turbulent layers inside the RL (Wehner et al., 2010), at the lower boundary of the nocturnal low-level jet (Siebert et al., 2007) and right above (Siebert et al., 2004), or inside (Platis et al., 2016) the the nocturnal inversion layer. The fresh-formed particles were assumed to mix later downwards.

Some environments might create special vertical distribution of NPF. Minguillon et al. (2015) measured the first 1 km of the vertical column over an urban area in Spain using a tethered balloon, and suggested that since high traffic pollution leads to a large condensation sink near ground, NPF could be enhanced at higher altitudes inside the PBL.

After the sunset, the stable nocturnal boundary layer starts to form above the ground, and the PBL above it transforms to a residual layer, which is not connected with the surface but can preserve the properties of the day's mixed layer. This means that the aerosol dynamics near the surface and in the residual layer can differ during the night. The organic vapor emissions and oxidation can continue after the sunset in the NBL and there the particles can grow in size (also seen between 18:00 and 24:00 on both 26 and 27 March in Fig. 2.2). In contrast, there is no persistent source for the condensable vapors in the residual layer, preventing further particle growth.

3 Measurement methods

3.1 Measurement sites

The aerosol measurements described in this thesis were performed at four measurement stations. One of them is located in southern Finland, while the three others are in Finnish or Swedish Lapland. Figure 3.1 shows the measurement sites on a map.

Värriö SMEAR I (Station for Measuring Ecosystem-Atmosphere Relations) station (Hari et al., 1994) was established in 1991 and it has provided long-term aerosol measurements since 1998. It is located in eastern Finnish Lapland (67^o 46'N, 29^o 37'E, 390 m above the sea level, a.s.l.) on top of a fell. There are several other fells around the site, the highest reaching up to 500–550 m a.s.l. The surroundings of the station is dominated by Scots pine (*Pinus sylvesteris*) forest, but also consists of wetlands and gorges. There are no local pollution sources, even the nearest small road is 8 km away from the station. The closest pollution sources are a mining area Kovdor, in Russia (45 km from the station), and three smelter industrial cities in Kola Peninsula, namely Nikel, Zapolyarnyj and Montchegorsk (150–200 km away from the station), also in Russia. **Papers I–II** used data collected from Värriö.

Pallas–Sodankylä GAW (Global Atmospheric Watch) station is located in the western Finnish Lapland. The atmospheric aerosol measurements are performed at Pallas, atop of Sammaltunturi fell (67^o 58'N, 24^o 07'E, 565 m a.s.l.) (Hatakka et al., 2003). The station is at the southmost part of an 50 km long fell area, where the tops reach to 600–800 m a.s.l. The area is mainly forested, but also consists of swamps and small lakes. The station is around 100 m above the timber line and the vegetation around it consists mainly of low vascular plants, moss and lichen. Below the timber line there is a mixed boreal forest. **Paper I** used data measured at Pallas.

Abisko research station is situated in the western Swedish Lapland. The aerosol measurements there are conducted at the Stordalen mire (68^o 21'N, 19^o 03'E, 360 m a.s.l.) (Svenningsson et al., 2008), 14 km east from the research station, and between lake Torneträsk and the Kiruna–Narvik road and railway. The area is inside a valley, where the vegetation consists of subarctic mire and birch forest. West from the station are the Scandinavian mountains, which peak above 1500 m a.s.l., and behind them at a distance of around 100 km, is the Atlantic Ocean. **Paper I** used data from Abisko.

Figure 3.1: A topographical map of Scandinavia showing the locations of the measurement stations. Map source: Natural Earth Data. Free vector and raster map data @ naturalearthdata.com.

Hyytiälä SMEAR II station (Hari and Kulmala, 2005), located in central Finland (62^o 51N, 24^o 17 E, 170 m a.s.l), provided the on-ground measurement support for the research flights described in **Papers III–V**. It is the most extensive station in the world to study the interactions between the biosphere and atmosphere. The surroundings of Hyytiälä are mainly cultivated boreal mixed forest of different ages, small lakes, farms and agricultural fields, and peatlands (e.g. Williams et al., 2011, see also Fig. 3.2b). This variety offers a possibility to study different biotopes and to combine the results with atmospheric research. Aerosol measurements conducted in Hyytiälä cover a wide range of physical, chemical and optical measurements, both in-situ and remote sensing, to characterize the aerosol processes from studies of the first steps of the nucleation and growth of secondary particles (Kulmala et al., 2012) all the way up to cloud condensation nuclei measurements (Sihto et al., 2011), and to the investigation of pollen particles (Manninen et al., 2014).

3.2 Aerosol number size distribution measurements

Differential mobility particle sizers (DMPS, Hoppel, 1978; Aalto et al., 2001) and *scanning mobility particle sizers* (SMPS, Wang and Flagan, 1990; Collins et al., 2002) were used to determine the aerosol number size distribution at the measurement sites or airborne. The basic principles of DMPS and SMPS are similar, but the running modes differ from each other. Both instruments first ensure that the particles have a known charge distribution and then separate and count the particles based on their different electrical mobilities, which are connected to their mass/charge ratios and which can be further converted to electrical equivalent mobility diameters.

In both DMPS and SMPS, the particles are brought to a known charge distribution by a radioactive source (e.g. ^{85}Kr or ^{14}C) and then classified by a differential mobility analyzer (DMA, Knutson and Whitby, 1975), which allows the particles having only a certain electrical mobility to pass through, based on DMAs classifying voltage. The SMPS goes through the electrical mobility and thus the diameter range with a continuous scan, whereas the DMPS uses a step-wise approach. The number of particles with a selected mobility diameter is measured by a condensation particle counter (CPC, e.g. McMurry, 2000), which condenses either butanol or water vapor onto the particles, and then detects them optically.

The size range of the SMPS and DMPS depends on the geometry and flow rates of the used DMA and the cut-off size of the CPC. A transfer function of the DMA is the probability that particles with a given mobility Z_p (which is inverse proportional to particle diameter D_p, Seinfeld and Pandis 2006) will pass the instrument (Knutson and Whitby, 1975). A high sheath flow Q_{sh} is needed for measuring small particles. The aerosol flow Q_a is chosen so that the diffusion losses (need for high Q_a) and the high resolution (need for low Q_a) are in balance. The ratio Q_a/Q_{sh} determines the width of the transfer function of the instrument and is usually between 1:5 and 1:20. Often, a twin-DMA system is used, where the size range is divided to two parts which are measured separately with two parallel DMAs with different flow rates, and counted with two CPCs. This allows one to measure both small and large particles with an improved performance. Twin-DMA systems are used at Värriö SMEAR I and Hyytiälä SMEAR II stations. Another way to optimize the detection at particles of both ends of the size range is to use a two-flow cycle, where the flows of the DMA are changed in the middle of the scanning cycle. The airborne SMPS used in **Papers III–V** measured

with the two-flow cycle with aerosol flow rates and sheath air rates of 1 lpm/5 lpm and 4 lpm/20 lpm for size ranges of 10–40 nm and 40–400 nm, respectively.

Earlier, the commercial butanol or water CPCs were limited in their techniques to detect only particles with a diameter of 3 nm or more (McMurry, 2000; Hermann et al., 2007; Mordas et al., 2008; Liu et al., 2006), and thus they could not detect the first steps of the NPF. In Airmodus *Particle size magnifier* (PSM, Vanhanen et al. 2011), the working fluid was changed to diethylene glycol (DEG), which has a lower saturation vapor pressure and higher surface tension than butanol (Iida et al., 2009). This makes it possible to detect particles down to 1 nm. DEG is condensed onto the particles until they are grown up to sizes of around 90 nm. After that the particles are not yet detectable with optical instruments, but they can be observed using a separate CPC. We tested PSM A09 and A10 in an aircraft to detect sub-3 nm particles, see next section.

CPCs and the PSM can detect both neutral and charged particles. To separate the neutral particles and ions, a Cessna light aircraft was equipped with a *Neutral cluster and Air Ion Spectrometer* (NAIS, Mirme and Mirme 2013; Manninen et al. 2016). It can detect both positive and negative ions with a diameter range of 0.8–42 nm, and neutral clusters with a diameter range between 2–42 nm (Manninen et al., 2011). The positive and negative ions are classified separately and simultaneously by two parallel DMAs. A series of 21 successive electrometers is connected to the DMAs to measure the ion currents, which are further converted into concentrations in size bins. In order to measure neutral particles, the aerosol particles are first charged by a corona needle charger, and then after filtering out the artifical corona ions, the particles are measured as ions. The NAIS runs in sequences of measuring the ion cluster, neutral cluster, and the background. The background mode detects the ions produced inside the instrument and the electrometer noise.

3.3 Aerosol measurements from aircraft as a part of extensive field campaigns

There are several ways to measure airborne aerosols in-situ in the lower atmosphere (Baumgardner et al., 2011). Many national or international organizations (such as Facility for Airborne Atmospheric Measurements, FAAM, in UK, Service des

Avion Français Instrumentès pour la Recherche en Environment, SAFIRE, in France, Deutschen Zentrum für Luft- und Raumfahrt, DLR, in Germany, or Earth Observing Laboratory's Research Aviation Facility, RAF, in USA) provide larger or smaller aircrafts dedicated to measurement flights. In Europe, the central network for airborne research is called EUropean Facility for Airborne Research (EUFAR) and it works for the cross-national access to national aircrafts and instruments (Formenti and Wendisch, 2008).

The airborne aerosol measurement platforms can vary from large commercial aircrafts (e.g. Petzold et al., 2015) to small unmanned multicopters (e.g. Brady et al., 2016). Large airplanes offer an option for heavy payload and extensive instrumentation with a wide spatial span and a possibility to high operation altitudes. Their disadvantages are, however, high operational costs, and high airspeeds, which reduces the spatial resolution. Platforms with moderate airspeed but capability to carry heavy instruments include the Zeppelin NT airship (Rosati et al., 2016; Manninen et al., 2013) or a pod carried by a helicopter (Siebert et al., 2006; Wehner et al., 2011, 2015). Additionally, tethered balloons (Spirig et al., 2004; Wehner et al., 2007; Minguillon et al., 2015; Moroni et al., 2015) and hot-air balloons (Laakso et al., 2007; Petäjä et al., 2012) have been used for measuring vertical profiles. Finally, the unmanned aircraft systems (UASs, also the term unmanned aerial vehicle, UAV, is used) are a rapidly developing platform type also for the aerosol measurements (Bates et al., 2013; Altstädter et al., 2015; Platis et al., 2016). They can also fly at very low altitudes. They often carry only light payloads and therefore their instrumentation is limited, although their capacity is extending. Depending on the UAS and the air traffic regulations, the UAS may need to be in the view of the operator even when using autopilot, their vertical span is often restricted to relatively low altitudes, and the radio link range can limit the horizontal span.

Paper III introduces a multi-platform measurement campaign "Biogenic Aerosols – Effects on Clouds and Climate (BAECC)" and the possibilities how to utilize its data. BAECC was a joint project between University of Helsinki (UHEL) and US Department of Energy's Atmospheric Radiation Measurements (ARM) program, and it took place between February and September 2014. The aim of BAECC was to better understand the role of biogenic aerosols in the microphysical properties of the clouds. BAECC brought together the extensive surface in-situ measurements conducted at the SMEAR II station, and remote sensing and in-situ observations of the tropospheric

(a) (b)

Figure 3.2: (a) Cessna 172FR aircrafts used as a platform ready to take-off at Tampere-Pirkkala airport on September 2014. (b) An aerial view to Hyytiälä SMEAR II station on March 2014. The surroundings of Hyytiälä consist of boreal forest, peatlands, small lakes, agricultured fields and few farms.

column. The campaign-wise measurements were supplemented with the 20-year-long comprehensive data from the SMEAR II station in the boreal environment. In-situ surface measurements included e.g. the gas phase precursors, and the physical, chemical and optical properties of aerosols, aerosol chemical composition, and precipitation and snow properties. The vertical column was measured in-situ by atmospheric soundings and aircraft-borne measurements, and remotely by radars and lidars. The flight measurements during BAECC were concentrated to three Intensive Observation Periods (IOPs) performed in March–April, May–June, and August–September. A total of 152 flight hours were flown with three aircrafts.

The aircraft-borne measurements described in **Papers III–V** were performed using Cessna FR172F type of light aircrafts as platform (Fig. 3.2a). They are four-seat single engine airplanes with unpressurized cabin. The operational cost is relatively low. During the measurement flights the crew consisted of two persons: a pilot and a scientific operator. The back seats were replaced by a rack for the instruments. Between 2009–2015 University of Helsinki performed 1–3 measurement flight campaigns per year, usually as parts of larger field campaigns. The aerosol instrumentation setup in the aircraft varied a little from campaign to campaign. The purpose was to measure the aerosol number concentration and the number size distribution, taking into account the limitations of the platform for the payload in terms of mass, size, and electricity consumption.

(a) (b)

Figure 3.3: (a) The airborne aerosol instrumentation consisted of an Airmodus Particle size magnifier (PSM) A10, TSI ultrafine condensation particle counter (uCPC) 3776, and scanning mobility particle sizer (SMPS). (b) Neutral cluster and Air Ion Spectrometer (NAIS) installed in an aircraft. Small picture shows the T-shaped inlet.

Until the year 2014, the inlet for the aerosol and gas instruments was mounted under the right wing and situated in the free airflow ahead of the leading edge of the wing. The inlet nozzle (Fig. 2.1) design followed the design presented in McNaughton et al. (2007). The air was directed into the cabin via a 4.2 m long steel tube and the instruments sampled from it via centerline sampling.

The particle number concentration during all campaigns was measured with a TSI ultrafine CPC (uCPC) model 3776. It recorded the concentration with a cut-off size of 3 nm and with 1-s time resolution. Since one of the main foci of the flights was on NPF and thus on nucleation mode particles, during the first campaigns the uCPC was complemented with two TSI 3772 CPCs with cut-off sizes tuned for 6 nm and 10 nm. This setup provided a two-bin size distribution for sub-10 nm particles with a 1-s time resolution, which corresponds to 35 m s^{-1} spatial resolution with 130 km h^{-1} air speed. This configuration was used in 2009 (part of the flights presented in **Paper IV**).

The TSI 3772 CPCs were replaced for campaigns in 2010 and later by a SMPS (for SMPS, see Sec. 3.2). The advantage of the SMPS was the possibility to measure the number size distribution between 10 and 350/400 nm with a dense diameter grid.

However, the drawback was a lower time resolution. One SMPS scan took two minutes, which corresponds to a horizontal resolution of 4.2 km with the velocity being 130 km h^{-1}.

The size range up to 400 nm covers the majority of the aerosol number concentration closure. Thus, the different cut-off sizes of the uCPC and the SMPS offer a way to calculate the number concentration of 3–10 nm particles, and this was done when calculating the regional vertical profiles in **Paper IV**. However, because the aerosol lifetimes in the atmosphere are shortest for the smallest particles (Williams et al., 2002), and therefore the expected variability of the smallest particles is the highest, the uncertainty of this 3–10 nm bin would be larger than the uncertainty of the size bins with larger particles. Additionally, in **Paper V** we investigated aerosol variation within the scale of a few kilometers, and averaging the difference of the uCPC and the SMPS values over the 2-min SMPS scans would have masked the small-scale concentration variation. As a consequence, this bin was not investigated in **Paper V**, but we concentrated on the total number concentration of particles and number size distribution of 10–400 nm particles.

The concentrations of carbon dioxide (CO_2) and water vapor (H_2O) were measured with a Li-Cor LI-840 gas analyzer. The basic meteorological parameters, such as temperature, relative humidity and static pressure were recorded. In addition, a GPS receiver located the position of the plane and a GoPro action camera was installed under the right wing to take pictures of the terrain, weather and clouds ahead every 15 seconds. All the instruments were modified to run with 12 VDC electricity supplied by accumulators. Fig. 3.3a shows the scientific instrumentation setup used in the May–June 2014 campaign.

As a part of the BAECC campaign (**Paper III**), airborne measurements by the Neutral cluster and Air Ion Spectrometer (NAIS) were performed in September 2014. During these measurements, two planes we used. The first one was equipped as described above. The second one, the 'NAIS plane', carried a NAIS modified for airborne measurements (Fig. 3.3b). This airborne NAIS was altered, for example, by adding chokes for the outflow to ensure a steady flow in conditions with changing inlet pressure. To increase the detection efficiency of the smallest particles and ions, the 4.2-m-long inlet line was replaced with a shorter sampling line. A T-shaped inlet with a 90-cm-long telescoped sample line was situated pointing outwards from the cabin's back window so that the T-shape was placed horizontally. This configuration was called a 'window

inlet'. The inlet was placed 22 cm under the wing, at a distance of 60 cm from the fuselage. The NAIS was mounted in a rack inside the cabin. The two planes were flown in a formation with a distance of about 40 m.

Starting from 2013, we tested Airmodus PSM A09 and A10 instruments to detect sub-3 nm particles. The instruments were modified to suit the airborne measurement by adding small pumps to create the vacuum and compressed air flows needed by the instruments. The aim was to calculate the concentration of 2–3 nm particles by subtracting the measured concentrations by the PSM and the uCPC, and correcting this with the diffusional tube losses. First, the PSM was placed in the rack with uCPC and SMPS, with the 4.2-m-long inlet line. However, the high tube losses of sub-3 nm particles via the main sample line together with the 0.7-m-long inlet line of the PSM (with inner diameter of 4 mm) were high (the transmission efficiency was 0.55 % for 2 nm particles through the main tube and the short tube, Brockmann 2001). This, combined with the high variation of the sub-3 nm particles along a flight path, made it hard to get reliable results for the concentration of 2–3 nm particles. In spring 2015, a short window inlet similar with that used with the NAIS in autumn 2014 was built for the other aerosol instruments. Also the configuration of PSM, uCPC and SMPS in the instrument rack was changed, so that the inlet lines for the PSM and uCPC were the shortest, and thus the tube losses were minimized. The setup proved to work, as both the NAIS and the PSM were found to detect sub-3nm particles (Leino et al., 2016).

Additionally, during the measurement campaigns in 2015, an Aircraft-Integrated Meteorological Measurement System (AIMMS-20, Aventech Research Inc.) with ARIM200 Digital Air Data Probe (ADP) was installed, tested and used. AIMMS-20 measured the three-dimensional air speed, basic meteorological parameters (temperature, relative humidity, pressure), and aircraft's location and position with 10 Hz time resolution. It consisted of a probe with pressure sensors, a relative humidity and temperature sensors; a module to measure the acceleration and angular acceleration; a two-antenna GPS system; and a central processing unit. Accurately measuring the atmospheric conditions with high time resolution together with aerosol number size distribution starting from 2 nm will improve the understanding of the NPF within the atmospheric turbulent processes, such as roll vortices (Lampilahti, 2016).

Aircraft-borne measurements are usually campaign-wise and cannot cover simultaneously the tropospheric column as a whole. In contrast, the lidars and radars operate usually continuously, but the interpretation of their data is complex and needs valida-

tion. **Paper III** showed that the airborne in-situ measurements by the aircrafts can be used to verify the remote sensing data. Also, the airborne measurements can provide input data for different atmospheric models or verify their results. Vice versa, models could help to interpret the temporally changing, multi-channel data that is measured by a three-dimensional moving platform.

4 Results

4.1 Particle growth by the vapors emitted from boreal forests

In **Paper I** we studied the net effect of dynamical aerosol processes on the aerosol population, when air masses travel hundreds of kilometers above a northern boreal environment. This analysis extended the study of Tunved et al. (2006), which showed that the net accumulated mass of the particles, produced by biogenic vapours emitted from boreal forests, is linearly dependent on the time the corresponding air mass spent over the boreal environment. Also, **Paper I** created a new method to study the changes in aerosol population using measurements from two stations.

The resulting change in the aerosol population by different aerosol processes, e.g. formation and condensational growth of particles, or removal by coagulation or deposition, depends on the vapor concentrations and meteorological conditions, and also on the initial aerosol population. To standardize the initial aerosol population states, two approaches were used. First, similarly as Tunved et al. (2006), we started from the assumption of fresh clean marine air, and investigated how the particle population develops after the air mass arrives over the continent and travels over land. We used the particle number size distribution measurements from three sites, namely Abisko, Pallas, and Värriö measurement stations in Swedish and Finnish Lapland.

To investigate the spatial history of the air masses, the HYSPLIT (HYbrid Single Particle Lagrangian Integrated Trajectory, Draxler and Hess 1998; Stein et al. 2015; Rolph 2016) model was used to calculate 96-hour backward trajectories. The HYSPLIT model simulates the atmospheric transport based on the Global Data Assimilation System (GDAS) reanalysis data (after 2004) and FNL Archive (before 2004) reanalysis data. The horizontal uncertainty of the trajectories is estimated to be 10–30 % of the travel distance (Stohl, 1998). Using the trajectories, the time the air masses had been over the continent before arriving at the stations, was calculated. Air masses originating from east or from too south were rejected from the analysis as being potentially too polluted. Then, the median particle number size distribution was calculated for each fixed time over land. The 'apparent' rates of growth of the particle diameter, particle number concentration, accumulated mass, and condensational sink were computed. We call these as 'apparent' rate, in order to separate them from the rates calculated when

measuring the particle dynamics, for example, within a single day at some measurement location.

The second approach used the suitable locations of the measurement stations. All the stations are located in the northern Scandinavia almost in a row, with around 200 km distance between Abisko–Pallas, and Pallas–Värriö (see map Fig. 3.1). We calculated the changes in the size distribution during the transport from one station to another. The HYSPLIT backward trajectory analysis was used to find those air masses that had passed both stations in a each pair. The number size distributions at the upwind station were clustered into mutually similar groups, and the net change in aerosol number size distribution for each of these cluster distribution during the travel was determined. The clustering algorithm used was *k-means* (Lloyd, 1982), which iteratively divides the data into groups having a maximal (chosen mathematical) distance between the different clusters and a minimal distance between the members of the clusters and the centers of the clusters. The algorithm is used to divide particle number size distributions to mutually similar groups (Beddows et al., 2009, 2014).

When looking at the median particle number size distributions as a function of the time the air masses had traveled over the land, all the measurement stations were observed to have similar patterns. The extended summer period, defined as 1 April–30 September, and the winter period defined as 1 October–31 March, differed clearly from each other (Fig. 4.1). In the northern boreal environment, the particle number concentrations during the summer period were found to increase at 'apparent' formation rates of about 0.01 cm^{-3} s^{-1} until the overland times of around 30–40 h and thereafter at rates between about 0.001 and 0.002 cm^{-3} s^{-1}. Eventually, the total particle number concentration saturated at 900–1100 cm^{-3}. This indicates that by the transport times of 30–40 h over land, the aerosol sources and sinks were in balance, and vapors did not anymore form new particles as the main process, but they were condensing onto the larger particles.

During the winter period, especially for Värriö and Pallas, the particle concentration increased slowly but smoothly as the function of time over land, although the particle diameters saturated to values of 45–60 nm. The biogenic emissions are the lowest during the winter (Hakola et al., 2009; Patokoski et al., 2014), and primary emissions are the highest (Karvosenoja et al., 2011; Lamberg et al., 2011) and thus we speculate that this slow increase in the particle number concentrations might be due to primary particle emissions.

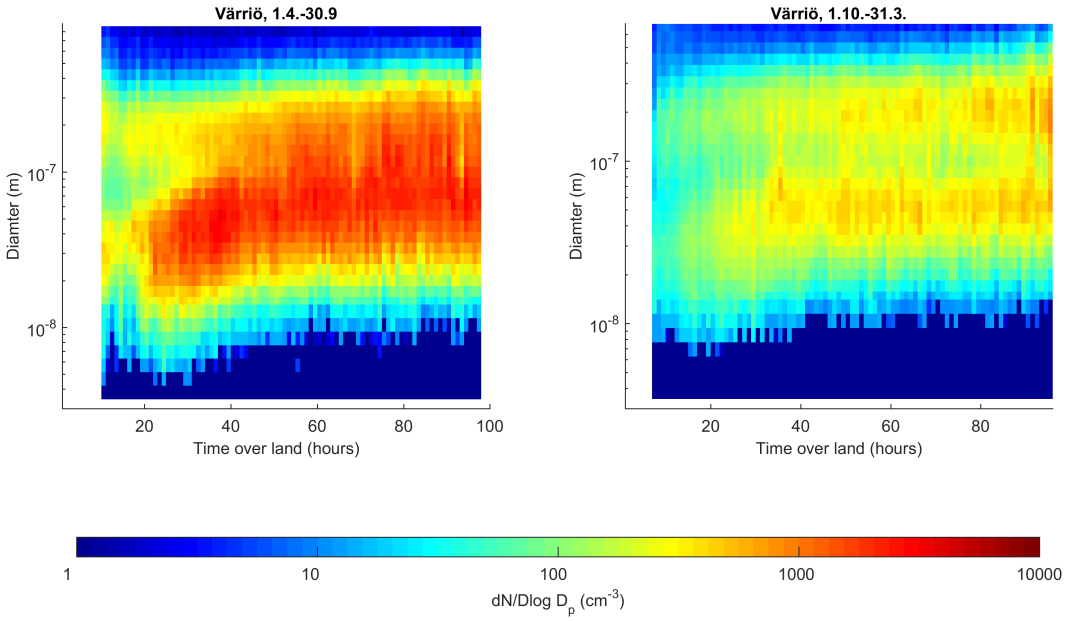

Figure 4.1: The median particle number size distribution at Värriö as a function of time the corresponding air mass has spent over land. In summertime, the biogenic vapours from the boreal forest participate in the nucleation and growth of the particles, which can be observed as increased particle concentration and growth of particle diameter.

We calculated also the 'apparent' increase rates for the mode peak diameter when air masses were passing over land. These 'apparent' growth rates were relatively similar for all the stations, with average values between 0.55 and 0.72 nm h^{-1} for size ranges of 12–80 nm (for Abisko) or 22–80 nm (for Pallas and Värriö). In contrast, the growth rates of the nucleated particles during the NPF events calculated in **Paper II** for Värriö in the summer months (Apr–Sep) during 1992–2011 were found to be, on average, in the range of 1.8–4.4 nm h^{-1}, and in **Paper I** the mean values (for all year data) between 2005–2008 were 3.7, 3.3, and 2.8 nm h^{-1} for Abisko, Pallas and Värriö, respectively. Consistent with these growth rates, it was found that when the air masses advected from west to east direction from one measurement site to another, the Aitken mode particles were found to increase in size with a rate of 0.6–1.3 nm h^{-1} (Fig. 4.2). The results are in line with e.g. Dal Maso et al. (2007) which showed an increased NPF probability for westernly air masses. When the air masses moved from east to west, the air was continental at the beginning, and had a larger aerosol loading and larger condensation sink, and therefore no such growth was observed.

Overall, the 'apparent' particle growth rates were clearly lower (by factors of 3–6) than the 'real' particle growth rates observed during the NPF event days. There are

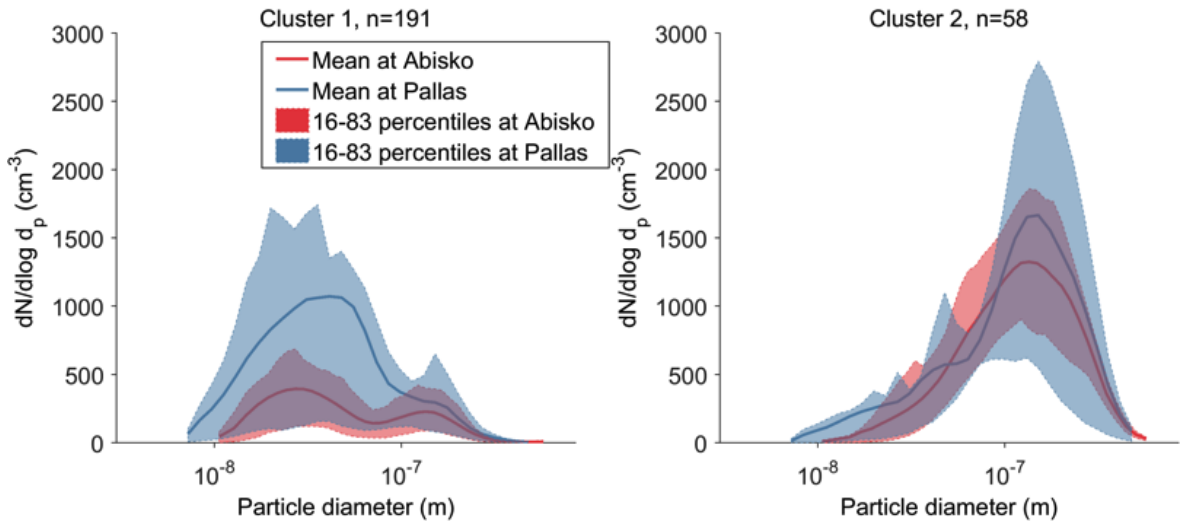

Figure 4.2: Changes in the particle number size distribution when the air masses were moving from Abisko to Pallas. First, the air masses that traveled from Abisko to Pallas were found using the trajectory analysis. The particle number size distributions at Abisko, at the moments when these air masses passed the station, were clustered. The initial particle number size distribution cluster at Abisko is marked by red color. Blue color shows how the particle number size distributions had evolved when arriving at Pallas. Solid curves are mean values and shaded areas show the 16th and 83th percentiles. The 'n' value refers to how many particle number size distributions were grouped to each cluster. The two largest clusters are presented here. The results showed that when the Aitken mode dominated at Abisko (left panel), on average the mode had grown in size with a rate of 1.3 nm h^{-1} and in concentration with a rate of 0.01 $cm^{-3}s^{-1}$ on the way to Pallas. When the accumulation mode dominated in Abisko (right panel) and on average the mode had grown in size with a rate of 1.3 nm h^{-1} and in concentration with a rate of 0.004 $cm^{-3}s^{-1}$.

several likely explanations for this. The 'real' particle growth is determined for the new particle formation event days and for sub-25 nm particles only. During the NPF days, the particle growth is, by definition, pronounced. Furthermore, the 'apparent' particle growth rate may be affected not only by particle growth but also by other aerosol processes affecting the shape of the number size distribution, such as size-dependent particle removal rate (Seinfeld and Pandis, 2006) or primary particle emissions.

4.2 The effect of reduced emissions at Kola Peninsula to the particle population at Värriö

The Värriö SMEAR I measurement station is located in eastern Lapland, and it is 150–200 km away from Kola Peninsula smelter industries, which are the largest anthropogenic sources of sulfur dioxide (SO_2) in the Arctic part of the Europe (Paatero et al., 2008). The emissions from Kola area, which include SO_2 and primary particles, have been reduced during the last decades due to the economic depression after the disintegration of the Soviet Union, but also due to cleaner ore material and recovery of SO_2 emissions for sulfur acid production (Symon, 2008). Concerning NPF, the SO_2 and particle emissions work in opposite directions: the diminishing of the primary particles reduces the condensation sink value, and thus eases the NPF, while the decreased SO_2 concentration hinders the sulfuric acid production and thus the formation of the new particles.

In **Paper II** a 14-year-long time-series of the particle number size distribution and SO_2 concentration measured at Värriö were used to investigate what is the net effect of the reduced emissions from Kola Peninsula on aerosols measured at Värriö. We used two methods to estimate the relative trends of various parameters between 1998 and 2011. The least-square covariance method took into account the seasonal cycle, and the non-seasonal Mann–Kendall -method was used to calculate Sen's slope with confidence intervals (Mann, 1945; Kendall, 1975).

The reduced emissions from Kola Peninsula were clearly seen on data. During the measurement period, the SO_2 concentration decreased with a yearly rate of approximate –11 % yr^{-1}. Since the direct sulfuric acid (H_2SO_4) measurements were lacking, the H_2SO_4 concentration was estimated using two proxies (Petäjä et al., 2009; Mikkonen et al., 2011) based on parametrization of the available measurements. The H_2SO_4 concentration proxies were found to decrease with rates between –5 % yr^{-1} and –9 % yr^{-1}. Similarly, the condensation sink decreased with a rate of –8 % yr^{-1}. These reductions in the SO_2 and H_2SO_4 proxies were high compared to values from similar study for Hyytiälä SMEAR II station in southern Center Finland (Nieminen et al., 2014). There the SO_2 concentration had decreased with a rate of –1.6 % yr^{-1}, the H_2SO_4 concentration proxy with a rate of –1.4 % yr^{-1}. As a result, the SO_2 and H_2SO_4 proxy concentrations between the stations were approaching each other.

The fraction of days when the NPF event occurred decreased with a rate of –3.4 % yr^{-1} during the observation period. The fraction of days with a most homogenous growth of the particles (event class I in classification based by Dal Maso et al. 2005) decreased most (–9.9 % yr^{-1}), whereas the fraction of the NPF events with inhomogeneous growth remained about constant during the measurement period. Thus, there were less nucleation events, and of these, a lower proportion was observed to have the pronounced smooth growth towards larger sizes. In addition, the fraction of the days when there was only growth of particles larger than 25 nm, or plumes of sub-25 nm particles but no continuous growth, remained stable. However, the average number concentration of nucleation mode particles, i.e. of particles with diameters of 3–25 nm, was found to increase with a yearly trend of around 4 %. One explanation for this increase is an particle production during the days with the irregular growth.

The air masses can advect hundreds of kilometers a day. The uniform and smooth growth of fresh nucleated particles observed at a fixed measurement station is thus associated with a large and regionally rather homogeneous air mass. The irregular NPF events, on the other hand, imply for non-uniform air masses. In Värriö the H_2SO_4 concentration explains 20–50 % of the growth of the new particles (**Paper II**), and the NPF event evolution can be interpreted from this viewpoint. When the sulfuric emissions from the smelters in Kola peninsula were high, the emissions were dispersed over a wide area and there was enough sulfur acid all over the area to start up a NPF events and to participate in the growth of particles. Then, frequent smooth NPF events were observed at Värriö. When the emissions were reduced, the fraction of the most smooth NPF event days when the initial steps of the nucleation was observed at Värriö, diminished. The fractions of days with more irregular growth or only growth of larger particles did not change, which indicates that the initial steps of the NPF occurs away from Värriö, within the area of higher H_2SO_4 concentration at closer to the smelters.

The NPF process can eventually produce particles that are large enough to act as CCN (Kerminen et al., 2012). In the boreal forest environment, the diameter limit for CCN particles is between 50–100 nm (Kerminen et al., 2005, 2012; Komppula et al., 2005; Sihto et al., 2011; Paramonov et al., 2013). Although the number concentration of nucleation mode particles increased during the studied 14 years, the number concentration of over 80 nm particles decreased with a rate around –4 % yr^{-1}. This can mean that the nucleated particles did not anymore grow to CCN sizes, which might have implications to the clouds and further to climate.

4.3 Vertical and horizontal extent of the NPF events

Papers IV and **V** present results from aircraft-borne aerosol measurements performed in the vicinity of the Hyytiälä SMEAR II station. The particle number concentration and number size distributions were measured. One aim of the measurements was to examine the horizontal and vertical extent of the NPF in the boreal environment both inside the PBL and in the FT.

In **Paper IV** three special cases from two seasons were studied. The first one was in early summer, 8 June 2009, and the second and third ones were in autumn, namely 21–22 September 2009, and 13 October 2010. All the case studies were the NPF event days. In the first case, the flight was flown at the same time as the start of the NPF was observed at Hyytiälä. In the second case, four flights with similar flight routes were conducted in order to to study the vertical diurnal pattern on a NPF event day. In the third case, a regional NPF was investigated both 100 km away from Hyytiälä just before the NPF was observed to start at ground, and near Hyytiälä four hours later. The flights suggested that the NPF occurs throughout the PBL, but not above it. Also, increased concentrations of nucleation mode particles were often detected in the upper part of the PBL.

The results in **Paper IV** indicated a high variability in the particle number concentration during the NPF events in the PBL. However, the flight patterns used in that paper, which consisted usually of two ascent–descent pairs flown over a large area with a constant vertical velocity, did not offer a possibility to distinguish between the vertical and horizontal variation. Because of this, one of the aims in **Paper V** was to characterize the airborne particle concentration and number size distribution variation inside the PBL with a horizontal scale of tens of kilometers during the NPF events, and to separate the horizontal variation from the vertical variation.

Paper V presents results from two spring time measurement campaigns, namely from May–June 2013 and March–April 2014. The spring seasons were chosen because then, on average, the NPF probability is higher than on the other seasons (Dal Maso et al., 2005; Nieminen et al., 2014). The meteorology in May-June 2013 was characterized by a warm three-week period with continental and polluted air masses originating from the east. As a result, high accumulation mode (80–400 nm) particle number concentration during the measurement flights were observed inside the PBL. As an opposite case, the air masses during the March–April 2014 originated mostly from the clean sector from

Arctic Ocean or Atlantic Ocean. This resulted in low condensation sink values which favored NPF in the PBL.

The NPF events are often found to be regional: inside the PBL they are observed over an area of hundreds of kilometers wide (Crumeyrolle et al. 2010; Wehner et al. 2007, **Paper IV**). Within these areas there can, however, be smaller-scale unhomogeneities in the particle formation. In **Paper V** we looked at the local scale (tens of kilometers) variation of the aerosol concentration during the flights during the NPF events. During one of the case study days, on 28 March 2014, an intense NPF event was observed at Hyytiälä. A research flight was conducted at the same time when the start of the event was observed in Hyytiälä. Before the nucleation was observed at the ground level, no sub-10 nm particles were observed by the aircraft instruments either. However, 30 minutes before the event was observed at the ground level, particles with diameter of 28 nm were observed in the residual layer. On the measurement day, the NPF event at the ground was observed to start by a burst of particles with diameters between 4–20 nm, which could indicate that when the rising PBL had reached the RL, the particles were mixed downwards simultaneously with the start of nucleation. Later, the airborne measurement showed that 40 minutes after the event started at ground, the nucleation mode particles were found throughout the PBL. The downward mixing of the fresh residual layer particles has been observed previously (Stratmann et al., 2003; Siebert et al., 2004, 2007; Wehner et al., 2010; Platis et al., 2016). Our case differs from these by the relatively large particle sizes of the RL aerosols. It is improbable that they had had enough time to form and grow up to almost 28 nm during the morning. Because of the suitable particle sizes, we speculate that these particles originated from a NPF event that took place in the previous day, and had remained in the residual layer throughout the night.

When NPF was observed to start at elevated atmospheric layers, there needs to be enough suitable precursor gases for nucleation in that layer. It can be speculated that the different precursor vapor concentrations in dissimilar environments could result in different vertical starting points of NPF. This would mean that the results measured in the more polluted Central European environment cannot be directly generalized to the northern boreal environment where, for example, the sulfur emissions are lower. The vertical profiles of the chemical composition of gases and clusters would help to interpret the nucleation mechanism.

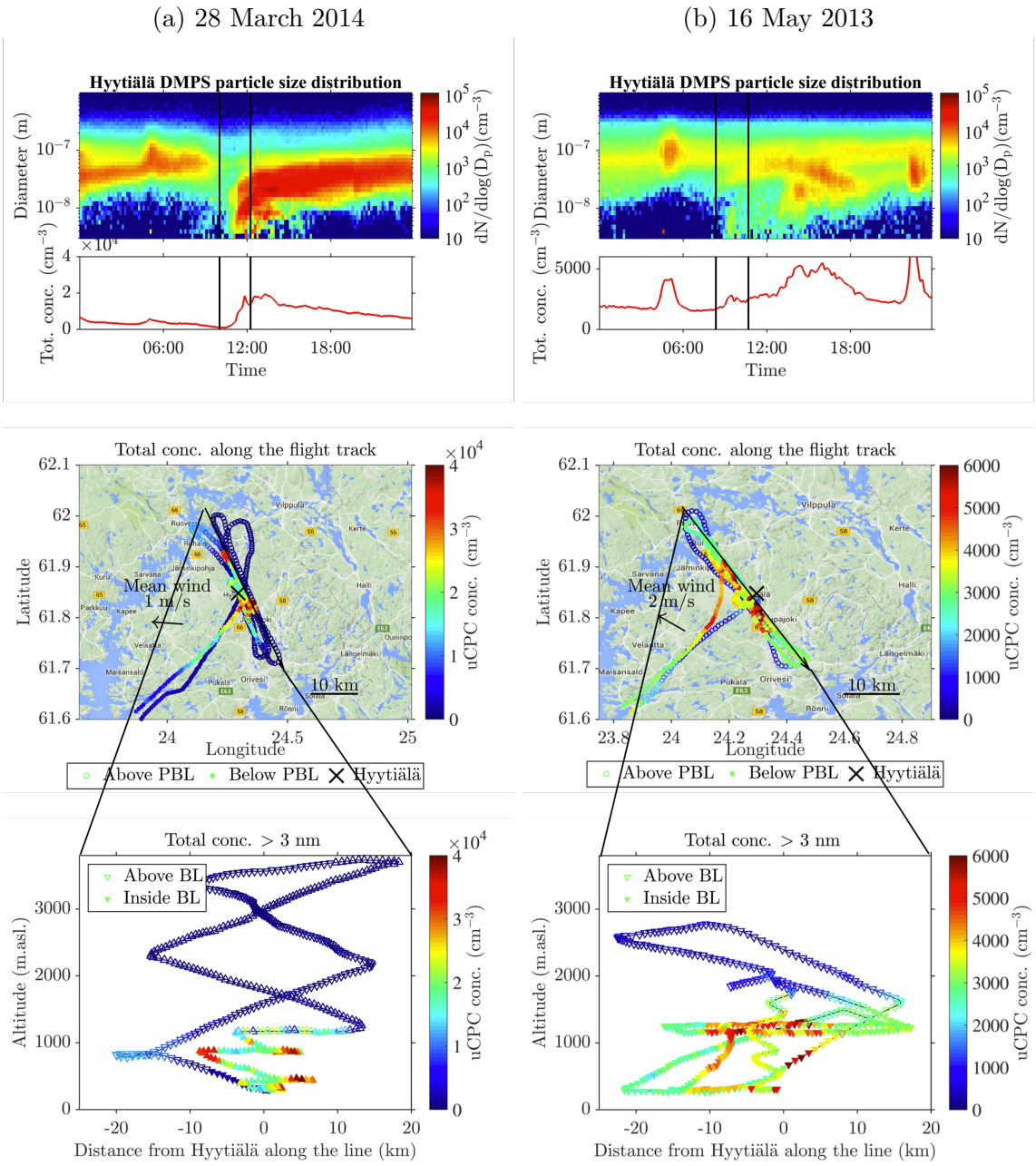

Figure 4.3: Airborne aerosol measurements in the vicinity of Hyytiälä during two NPF days. The uppermost panels show the Hyytiälä DMPS size distribution, the flight time is marked by two vertical lines. The middle panels show the total number concentration plotted ontop of a map. For the bottom panels the flight track on the map is reduced to a line. X-axis shows the distance from Hyytiälä along this line, y-axis is the altitude and color show again the total number concentration.

The flight legs inside the PBL formed parallel lines at different altitudes. The analysis revealed areas with intensified sub-10 nm particle concentration through the PBL (Fig. 4.3a). During the morning flight these areas were also more humid than their surroundings, which led us to made a hypothesis that together with the water vapor from vegetation and soil, the updrafts could carry suitable precursors to participate in the nucleation. However, during the afternoon the mixing had smoothened the relative concentration differences: whereas in the morning the concentration differences between these areas with the enhanced NPF and the areas around them had four-fold difference, in the afternoon it was only 1.5-fold. This suggest that, at in least some cases, when the particles grow to CCN size the initial spatial variability in the NPF was smoothed out.

On 16 May 2013 an uncontinuous growth of sub-25 nm particles was observed at Hyytiälä (Fig. 4.3b), and a high spatial variation in the particle number concentration was observed during both morning and afternoon flights. The lengths of the areas with mutually different concentrations varied from a few kilometers to over ten kilometers. The airborne measurements showed also vertical variation inside the PBL, but it was not as intense as the horizontal variation.

The instrumentation of the aircraft lacked the chemical composition measurements for gases or particles. Also the airborne turbulence measurements were not yet started in 2014. Thus we were not able to explain the atmospheric conditions reliably neither the basis for the high variability of the particle number concentrations or the areas with the intensified NPF in detail. The variation may be connected to the land use and surface properties, or meteorological phenomena.

The ceiling altitude of the research flights was usually between 2 and 3.8 km, so the transition between the PBL and free troposphere (FT), as well as the lowest parts of the FT could be studied. We found that the NPF events seen on ground were limited inside the PBL (**Paper IV-V**). This is in line with previous airborne NPF observations (e.g. Laakso et al., 2007; O'Dowd et al., 2009; Crumeyrolle et al., 2010).

It was found that separate to the NPF inside the PBL, the NPF in the FT was a frequent phenomena: sub-25 nm particles were observed inside the FT during all the three case study flights in **Paper IV**, and on 9 out of 27 of the flight days with an ascent at least up to 2 km during the 2013 campaign, and on 7 out of 10 of the flight days during the 2014 campaign in **Paper IV**. Also, during several days in 2014, we

were able to follow the growth of particles between the morning and afternoon flights. In the majority of these cases, the NPF in the FT was restricted to a certain altitude, not covering for example all the measured FT altitudes between 1.5 km and 3.5 km. When investigating the backward trajectories of the air masses of the corresponding altitudes, we found that in the majority of the cases, the air masses had been lifted up from the PBL 0.5–3 days before they were observed. One possibility is that the air masses containing vapors originating from ground or ocean had been oxidized during the transport in the FT. According to Bianchi et al. (2016), in Central Europe oxidation processes in the FT required 1–2 days to produce enough condensable vapors that the NPF can happen. It is plausible that lower vapor concentration or lower solar radiation can lengthen this time in Northern Europe. The confirmation of these hypotheses would require airborne precursor gas measurements together with aerosol chemical composition measurements.

4.4 Representativeness of the Hyytiälä on-ground aerosol measurements

In **Paper V** we investigated the representativeness of Hyytiälä SMEAR II data during the measurement campaigns. Overall, the median difference between the simultaneously measured airborne and Hyytiälä number concentration values was low. An intercomparison between Hyytiälä DMPS and airborne SMPS and uCPC was performed after the May–June 2013 campaign. Three-day side-by-side measurements showed that the instruments differed, on average, by less than 10 %, which is a good agreement. The area around Hyytiälä was divided into concentric 'shells' separated by 5 km, and for each of these distance bins the median difference between the simultaneous concentration measurements of on-ground at SMEAR II and airborne was calculated. When looking at the number concentration of particles larger than 3 nm, these median differences showed that airborne concentrations were 0–17 % larger than the on-ground values. This is only slightly larger than the median difference between the instruments during the intercomparison. The median difference was larger (6–82 %) for nucleation mode (10–25 nm) particles, and smaller (between –2 and +6 %) for CCN size particles (80–400 nm). Larger particles have longer lifetimes and are thus mixed better in the PBL, which explains their smaller difference.

The vertical aerosol concentration profiles were calculated separately for both springs. Figure 4.4 shows the day time median vertical profiles for the total concentrations of particles over 3 nm and for the size bins of 10–25 nm and 80–400 nm. The springs differed from each other. Three processes were found to cause the differences. During 2014, a high pressure prevailed during many of the measurement days. It resulted in a large-scale subside and the mixing between the PBL and the free troposphere was hindered (similarly as in Hamburger et al., 2011). Also, as seen from Fig. 2.4, the range of the PBL height varied more during 2013 than during 2014, particularly in the mornings. This caused a smoother vertical median concentration profile for 2013, since single altitude bins could contain particle concentration values both within and above the ML. Finally, during the 2014 campaign, a majority of the measurement days were clear NPF event days, whereas during the 2013 campaign days, irregular particle growth dominated. This was seen as larger total concentrations and larger nucleation mode particle concentrations inside the PBL during 2014 spring than during 2013 spring.

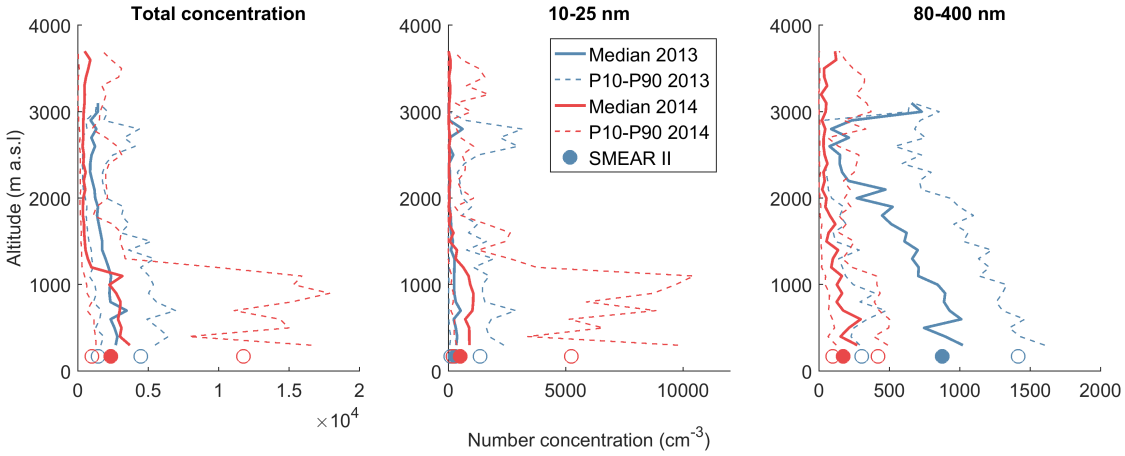

Figure 4.4: The median vertical profiles for total particle concentration, and concentrations of the size bins 10–25 nm and 80–400 nm. The daytime data (between 08–16) is considered.

5 Review of papers and the author's contribution

This thesis contains five papers. I am solely responsible for the introduction of this thesis.

Paper I reported the investigations on the changes on particle number size distribution over time periods from hours to several days. Particle number size distributions from three sub-arctic measurement sites were used. A NPF analysis was performed. The particle number size distributions were analyzed as a function of the time that the air masses had travelled over land, and additionally the changes in the particle size number distribution when air masses passed two stations were quantified. On average, particles were found to grow in size by vapor condensation when the air masses advected over boreal environment. *Author's contribution*: I analyzed the particle and trajectory data, developed the method to calculate the changes between two stations and wrote most of the article.

Paper II investigated the effect of the Kola Peninsula's SO_2 emissions on NPF observed in eastern Finnish Lapland. 14-year-long data sets of particle number size distribution, H_2SO_4 proxy and air mass backward trajectories showed that the reduction of SO_2 from Kola had resulted in fewer observed NPF days, and lower number concentration of CCN size particles in Värriö. *Author's contribution:* I did the backward trajectory analysis and wrote part of the text.

Paper III described an overview of an extensive field campaign conducted in 2014 at the Hyytiälä SMEAR II station. The main focus of the campaign was aerosol–cloud interactions. Flight measurements were performed during three Intensive Observation Periods (IOPs) between March and September. *Author's contribution*: I performed the majority of the Cessna flight campaign planning, airborne instrument preparations, and around one third of the measurement flights. I processed the airborne raw data to the final format. I wrote of the airborne measurements for the paper and participated in the writing process.

Paper IV studied NPF experimentally in the lower troposphere. The measurements confirmed that the NPF events were spatially over a hundred kilometers wide. The vertical profiles showed an enhancement of the nucleation mode particles in the upper part of the PBL in the beginning of the NPF event. *Author's contribution*: I participated in the flight campaign preparation and operated around half of the measurement

flights in 2010, and analyzed the 2010 case study. I wrote the corresponding part of the text.

Paper V combined results from two airborne campaigns performed in 2013 and 2014 in the vicinity of the Hyytiälä SMEAR II station. Median vertical profiles of aerosols in the lower troposphere were presented The aerosol number concentrations measured at Hyytiälä were found to, on average, represent well its surroundings. Sample case studies showed local concentration variations during NPF. *Author's contribution*: I did a large part of the flight campaign preparations in 2013, and the majority of them in 2014. I performed around half of the flight measurements in 2013, and around one third of them in 2014. I processed the raw data to the final format and analyzed the data. I wrote most of the paper.

6 Conclusions

The focus of this thesis is to understand the natural and anthropogenic processes that alter the particle number size distribution in the boreal environment. Boreal forests cover 13.7 million km^2 in Fennoscandinavia, Russia and Canada, which is around one third of Earth's forested areas (Grace, 2004), and thus understanding their impact to the aerosol particle load is important. In **Papers I-II** we studied aerosol populations and their dynamics based on measurements at three ground stations located in Finnish and Swedish Lapland. The distances between the stations, and from major anthropogenic source areas to one of these stations, were hundreds of kilometers, which makes it possible to study the regional dynamics of aerosol number size distribution. In **Papers III-V** the vertical profiles of the aerosol number size distributions were investigated together with the planetary boundary layer behavior within a local span (tens of kilometers) of a measurement station situated in southern Central Finland. The main results of this thesis are the following:

1) Vapors emitted from boreal forest are an important contributor to aerosol growth, even when explicit new particle formation is not observed.
Aerosol number size distribution data from three measurement stations within distances of hundreds of kilometers from each other was used to to determine the net change in the aerosol number size distribution when an air mass travels over boreal environment. During the growing season, there was a continuous flow of condensing mass onto the Aitken mode particles. This could be seen in that the longer the airmass had been over continent, the larger the particles were in diameter and mass. This accumulated mass load was probably due to vapors that had been emitted from vegetation and oxidized in the atmosphere to have lower volatility. The Lapland area in Finland and Sweden was found to be homogenous in this sense, as the observed 'apparent' growth rates were similar at all three studied stations.

Also, we developed a new method to measure the changes in the aerosol population based on measurements performed at two sites within a distance of tens or hundreds of kilometers. Using the air mass trajectory analysis, air masses traveling over two stations could be identified. The different initial aerosol size distributions were clustered to mutually similar groups, and the changes in each of these groups were studied separately. The analysis of particles arriving from the ocean to the continent includes an assumption of particle free air above ocean. The new method is suitable for any

geographical location with two or more measurement stations situated relatively close to each other.

2) Quantification of the effect of Kola Peninsula sulfur emission reduction on aerosols observed in Eastern Finnish Lapland.
The anthropogenic SO_2 emission from Kola Peninsula smelter industries are the largest or the second largest in the Arctic region (Benkovitz et al., 1996; Prank et al., 2010). Their impact touches an area that has a radius of at least few hundreds of kilometers. The economical depression after the collapse of the Soviet Union, changes in the ore composition, and the increased recovery of the SO_2 from the processes have led to a decrease of SO_2 pollution in the Kola Peninsula during the past 25 years. In **Paper II** the impacts of this reduction on aerosols observed in Eastern Finnish Lapland, around 200 km away from the smelters, were quantified. The emissions were found to influence on the observed NPF events in Värriö, as when the air masses passed the Kola Peninsula, the NPF was more frequent compared with the other directions. The results showed that although there was a growing trend of the concentration of the nucleation mode particles during the 14-year-long research period, the observed smooth NPF events reduced by almost 10 % yr^{-1} and the concentration of CCN sized particles decreased. This indicates that the particles did not grow as often to climatically relevant sizes as before, and also emphasizes the results from **Paper I** that in northern boreal environment the condensation growth of the particles is partly invisible, if looking only the days when a 'banana curve' can be observed from DMPS plot.

3) Combining in-situ measurements both on-ground and airborne with remote sensing methods offers a way to understand the atmospheric processes.
The surface-based measurements offer limited possibilities to explain the interactions between the aerosols and the atmospheric processes in the planetary boundary layer. In **Paper III** we introduced an instrumentation setup to in-situ measure atmospheric aerosols in the lower troposphere and implemented airborne measurement campaigns in the vicinity of the Hyytiälä SMEAR II station. The data gained adds the comprehensive understanding on the chain of aerosol processes starting from biogenic emissions, continuing with the nucleation of new particles, their growth, and ending up to aerosol–cloud interactions.

4) The NPF events were observed to cover an area of over a hundred kilometer, but when the newly formed particles were growing, there were horizontal differences in the aerosol number concentration within spatial scales

of kilometers to tens of kilometers. These horizontal concentration variances within kilometers scale were larger than the vertical concentration gradient in the planetary boundary layer.
Results from six flight campaigns to measure the particle number size distribution in the lower troposphere were presented. Due to different flight profile schemes, the flights in **Paper IV** studied the regional extend of the NPF, and the flights in **Paper V** the local scale variability in the vicinity of Hyytiälä SMEAR II station. The flights that overlaid a larger area showed that inside the PBL, the NPF was occurring over an area of over a hundred kilometer wide. However, particularly in the beginning of the NPF events there were areas with intensified particle formation and up to fourfold number concentration increases compared to their surrounding areas. In some cases these areas were connected to larger water vapor concentrations than the surrounding areas, which could indicate updrafts from land. However, since neither the turbulence nor chemical composition of the precursor vapors were measured, the exact mechanism behind these differences could not be concluded.

5) Weeks-long airborne datasets from two spring season campaigns showed that, on average, the aerosol number size concentration measured at on-ground boreal forest station represents the aerosol concentration well within a scale of tens of kilometers.
We quantified the median number concentration difference between the airborne measurements inside the planetary boundary layer and at an on-ground station. For total concentration, the airborne values were 0–17 % larger than the on-ground concentrations. The difference was larger for smaller (10–25 nm) particles as expected because of their shorter lifetime.

The data sets presented in **Papers III** and **V** (partly same) are extensive airborne aerosol measurement data sets. They show that similarly than at ground level measurements (Dal Maso et al., 2005; Nieminen et al., 2014) the aerosol properties and dynamics in the planetary boundary layer and above it depend in a very complicated way on the meteorological conditions, emissions and the air mass history. Short flight measurement campaigns are therefore not sufficient to create comprehensive understanding of the vertical extent.

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